

On exciton decoherence in quantum dots

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Abstract: The effects resulting due to dressing of an exciton with phonons are analysed as the source of unavoidable decoherence of orbital degrees of freedom in quantum dots. The dressing with longitudinal optical phonons results in energetic shift of order of a few meV even of the ground state of exciton in a state-of-the-art InAs/GaAs dot and the mediating role of longitudinal acoustical phonons is essential in this process. The characteristic time needed for dressing of the exciton with optical phonons is of a picosecond order. That time can be regarded as the lower limit for decoherence for optically driven quantum gates employing self-assembled quantum dot structures.

Keywords: quantum dots, exciton-phonon interaction, polaron, decoherence

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1 Introduction

We consider an exciton, i.e. an electron–hole pair created in a quantum dot (QD) by extremely short optical pulse (Borri 2001, Kikkawa 1988). The rapidly created bare exciton is further dressing with phonons, which results in arising a composite quasiparticle being the coherent mixture of the electron–hole pair and phonons. Its energy is lower than the bare exciton energy. In the case of interaction with only optical phonons this composite particle can be called exciton–polaron in analogy to the Frolich electron–polaron. In a realistic model much weaker deformation effects responsible for the interaction with acoustical phonons are included.

The observation of the exciton dressing with phonons is possible when the dipole coupling with the electromagnetic field is sufficiently strong to preserve a non adiabatic (i.e. rapid) exciton creation (cf. 0.2ps laser pulse in Ref. (Borri 2001)) much faster than dressing with phonons (of ps scale as we will show below). We consider a weakly polar medium for QDs (GaAs) in which the interaction of electrons with longitudinal optical (LO) phonons is dominating in terms of the influence on the quasiparticle energy shift. Much weaker interaction with gap-less longitudinal acoustic (LA) phonons plays an important role in kinematics of the dressing process, being a channel of energy transfer from exciton. In order to determine the limit of decoherence time of the optical excitations in QDs, this LA channel for dressing exciton with LO/LA phonons must be included.

We describe and analyse the dressing and the relaxation of exciton problems in detail for typical self-assembled strain induced InAs/GaAs QDs (Jacak 1998) within the Green function formulation. We model the QD by the parabolic confinement potential.

2 Phonon dressing description

In order to investigate the time evolution of the lowest exciton state, we consider the Hamiltonian describing a single electron and a hole interacting with phonons

$$\begin{aligned} \mathcal{H} = & \mathcal{H}_e(\mathbf{r}_e) + \mathcal{H}_h(\mathbf{r}_h) - \frac{e^2}{\epsilon_0 |\mathbf{r}_e - \mathbf{r}_h|} + \sum_{\mathbf{k}s} \hbar \omega_{\mathbf{k}s} b_{\mathbf{k}s}^\dagger b_{\mathbf{k}s} - \frac{e}{N^{1/2}} \left(\frac{2\pi \hbar \Omega}{v \tilde{\epsilon}} \right)^{1/2} \sum_{\mathbf{k}} \frac{1}{k} \\ & \times (b_{\mathbf{k}o} + b_{-\mathbf{k}o}^\dagger) (e^{i\mathbf{k}\mathbf{r}_e} - e^{i\mathbf{k}\mathbf{r}_h}) - \frac{1}{N^{1/2}} \left(\frac{\hbar}{2Mc_a} \right)^{1/2} \sum_{\mathbf{k}} k^{1/2} (b_{\mathbf{k}a} + b_{-\mathbf{k}a}^\dagger) (\sigma_e e^{i\mathbf{k}\mathbf{r}_e} - \sigma_h e^{i\mathbf{k}\mathbf{r}_h}). \end{aligned} \quad (1)$$

Here $b_{\mathbf{k}s}^\dagger$ ($b_{\mathbf{k}s}$) denote the phonon creation (annihilation) operators, ($s = o$ for LO, $s = a$ for LA), Ω denotes the frequency of the zero wave vector LO phonons, c_a – the LA phonon frequency, $\sigma_{e,h}$ – deformation constants for the electron and hole, M – the mass of ions in the elementary cell, v – the elementary cell volume, N – the number of cells in crystal, $\tilde{\epsilon} = (1/\epsilon_\infty - 1/\epsilon_0)^{-1}$ – the effective dielectric constant. The electron (hole) part of the Hamiltonian

$$\mathcal{H}_i(\mathbf{r}_i) = -\frac{\hbar^2}{2m_i^*} \nabla_i^2 + \frac{1}{2} m_i^* (\omega_0^i)^2 r_{\perp i}^2 + \frac{1}{2} m_i^* (\omega_z^i)^2 z_i^2 \quad (2)$$

(where $i = e, h$, $r_{\perp i}^2 = x_i^2 + y_i^2$) contains the vertical and horizontal confinement potentials. There are characteristic confinement sizes $l_i = \sqrt{\frac{\hbar}{m_i^* \omega_0^i}}$, $l_{iz} = \sqrt{\frac{\hbar}{m_i^* \omega_z^i}}$ ($\omega_0^i \ll \omega_z^i$). Finding

the eigenfunctions $\Phi_n(\mathbf{r}_e, \mathbf{r}_h)$ of the exciton Hamiltonian

$$\mathcal{H}_{ex} = \mathcal{H}_e(\mathbf{r}_e) + \mathcal{H}_h(\mathbf{r}_h) - \frac{e^2}{\epsilon_0 |\mathbf{r}_e - \mathbf{r}_h|}, \quad (3)$$

we write the Hamiltonian (1) in the second quantisation representation in the form of the sum of unperturbed and interaction parts

$$\mathcal{H} = \sum_n \hbar E_n a_n^\dagger a_n + \sum_{\mathbf{k}, s} \hbar \omega_{\mathbf{k}s} b_{\mathbf{k}s}^\dagger b_{\mathbf{k}s} + \frac{1}{N^{1/2}} \sum_{n_1, n_2, \mathbf{k}, s} F_s(n_1, n_2, \mathbf{k}) a_{n_1}^\dagger a_{n_2} (b_{\mathbf{k}0} + b_{-\mathbf{k}0}^\dagger), \quad (4)$$

where

$$F_o(n_1, n_2, \mathbf{k}) = -\frac{e}{k} \left(\frac{2\pi \hbar \Omega}{v \tilde{\epsilon}} \right)^{1/2} \int \Phi_{n_1}^*(\mathbf{r}_e, \mathbf{r}_h) (e^{i\mathbf{k}\mathbf{r}_e} - e^{i\mathbf{k}\mathbf{r}_h}) \Phi_{n_2}(\mathbf{r}_e, \mathbf{r}_h) d^3\mathbf{r}_e d^3\mathbf{r}_h, \\ F_a(n_1, n_2, \mathbf{k}) = -\left(\frac{\hbar k}{2Mc_a} \right)^{1/2} \int \Phi_{n_1}^*(\mathbf{r}_e, \mathbf{r}_h) (\sigma_e e^{i\mathbf{k}\mathbf{r}_e} - \sigma_h e^{i\mathbf{k}\mathbf{r}_h}) \Phi_{n_2}(\mathbf{r}_e, \mathbf{r}_h) d^3\mathbf{r}_e d^3\mathbf{r}_h. \quad (5)$$

The linear response of the exciton to the electromagnetic field is described by using the retarded Green function of the electric current operators

$$\mathbf{j}(t) \sim \sum_n \mathbf{d}_n [a_n(t) + a_n^\dagger(t)], \quad (6)$$

here \mathbf{d}_n denotes an effective dipole moment of the exciton (Elliott 1957, Mahan 2000). The spectral density of the linear response function is determined by the causal one-particle functions $G_{nn'}(t) = -i\langle T\{a_n(t)a_{n'}^\dagger\} \rangle$ (where the average is taken with Hamiltonian (4)) which we evaluate solving the Dyson equation. We calculate the spectral function $A(\omega) \equiv -2\text{Im}G_{00}^r(\omega) = -2\text{Im}G_{00}(\omega + i0^+)$ and its time course $A(t) = (2\pi)^{-1} \int_{-\infty}^{\infty} A(\omega) e^{-i\omega t} d\omega$. In order to determine the one-particle causal function, we evaluate the components of the mass operator using standard method of the solution of the Green function equations of motion

$$\left(i\hbar \frac{d}{dt} - E_n \right) G_{nn'}(t) - \frac{1}{N^{1/2}} \sum_{n_1, \mathbf{k}s} F_s(n, n_1, \mathbf{k}) R_{n_1 n'}(\mathbf{k}s, t) = \delta(t) \delta_{nn'}, \quad (7)$$

where

$$R_{n_1 n'}(\mathbf{k}s, t) = -\frac{i}{\hbar} \langle T\{a_{n_1}(t)[b_{\mathbf{k}s}(t) + b_{-\mathbf{k}s}^\dagger(t)]a_{n'}^\dagger(0)\} \rangle, \quad (8)$$

with additional condition $\langle \tilde{\varphi}_{\mathbf{k}s}(t) \rangle \equiv \langle b_{\mathbf{k}s}(t) + b_{-\mathbf{k}s}^\dagger(t) \rangle = 0$ (Martin 1959, Engelsberg 1963). Its Fourier transform takes the form of the Dyson equation

$$G_{nn'}(\omega) = G_{nn_1}^{(0)}(\omega) + \sum_{n_1 n_2} G_{nn_1}^{(0)}(\omega) \Sigma_{n_1 n_2}(\omega) G_{n_2 n'}(\omega). \quad (9)$$

Neglecting the off diagonal Green functions, one finds the diagonal elements of the mass operator

$$\Sigma_{nn}(\omega) = \frac{1}{N} \sum_{\mathbf{k}s} \sum_{n_1} F_s(n, n_1, \mathbf{k}) F_s(n_1, n, -\mathbf{k}) \left\{ \frac{\Gamma_{n_1 n, \mathbf{k}s}(\omega - \omega_{\mathbf{k}s}, -\omega_{\mathbf{k}s}) [1 + n(\hbar \omega_{\mathbf{k}s})]}{\hbar(\omega - \omega_{\mathbf{k}s}) - E_{n_1} - \Sigma_{n_1 n_1}(\omega - \omega_{\mathbf{k}s})} \right. \\ \left. + \frac{\Gamma_{n_1 n, \mathbf{k}s}(\omega + \omega_{\mathbf{k}s}, \omega_{\mathbf{k}s}) n(\hbar \omega_{\mathbf{k}s})}{\hbar(\omega + \omega_{\mathbf{k}s}) - E_{n_1} - \Sigma_{n_1 n_1}(\omega + \omega_{\mathbf{k}s})} \right\}. \quad (10)$$

Here $n(x) = (e^{x/T} - 1)^{-1}$, and $\Gamma_{nn',\mathbf{k}s}(\omega_1, \omega_2)$ denotes the Fourier transform of a vertex function

$$\Gamma_{nn',\mathbf{k}s}(t, t') \equiv -\frac{\delta G_{nn'}^{-1}(t)}{F_s(n, n', \mathbf{k})\delta\tilde{\varphi}_{\mathbf{k}s}(t')}. \quad (11)$$

satisfying $\Gamma_{nn,\mathbf{k}s}(\omega, \omega_1) \approx 1 - \hbar^{-1}d\Sigma_{nn}/d\omega$ when consider terms of the lowest order in \mathbf{k} . Let us denote real and imaginary part of the mass operator, respectively, $\text{Re}\Sigma_{00}(\omega) \equiv \Delta(\omega)$, $\text{Im}\Sigma_{00}(\omega) \equiv -\gamma(\omega)$.

We may solve the mass operator equation (10) with the assumption that the dressed exciton is a durable composite particle which is undamped (Suna 1964). It corresponds to the strictly solvable one level model of (Krummheuer 2002). We expect in this case that the exciton Green function has a real pole at frequency corresponding to the exciton–polaron energy. In the frequency region in vicinity of this pole, the imaginary part of the mass operator can be neglected compared to the real part, which allows us to write the self-consistent equation for $\Delta(\omega)$ (assuming $\Delta(\omega) \simeq -\Delta = \text{const}$)

$$-\Delta \simeq \frac{\hbar}{N} \sum_{\mathbf{k}} J_{00}^o(\mathbf{k}) \left\{ \frac{[1 + n(\hbar\omega_{\mathbf{k}o})]}{E_0 - \tilde{E}_n - \hbar\omega_{\mathbf{k}o}} + \frac{n(\hbar\omega_{\mathbf{k}o})}{E_0 - \tilde{E}_n + \hbar\omega_{\mathbf{k}o}} \right\} \quad (12)$$

which may be solved exactly ($\tilde{E}_n = E_0 - \Delta$, $J_{nn'}^s(\mathbf{k}) = F_s(n, n', \mathbf{k})F_s(n', n, -\mathbf{k})$). The energy shift due to the interaction with LA phonons is negligible. One can calculate the form factors

$$\begin{aligned} J_{00}^o(\mathbf{k}) &= \frac{\pi e^2 \hbar \Omega}{18 v \tilde{\epsilon}} (L_e^2 - L_h^2)^2 e^{-l_{\perp} k_{\perp}^2 / 2 - l_z k_z^2 / 2}, \\ J_{00}^a(\mathbf{k}) &= \frac{\hbar k}{2 M c_a} (\sigma_e - \sigma_h)^2 e^{-l_{\perp} k_{\perp}^2 / 2 - l_z k_z^2 / 2}, \end{aligned} \quad (13)$$

(here $l = l_e \approx l_h$, $l_z = l_{ze} \approx l_{zh}$, $L_e = \sqrt{\frac{\hbar}{m_e^* \omega_m}}$, $L_h = \sqrt{\frac{\hbar}{m_h^* \omega_m}}$, and $\omega_m = \sqrt{\frac{m_e^*}{m}(\omega_0^e)^2 + \frac{m_h^*}{m}(\omega_0^h)^2}$, $m = m_e^* + m_h^*$) (Davydov 1976).

However, we intend to describe the dressed exciton as a damped quasiparticle at non zero temperature range. When include many exciton energy levels, the damping is a consequence of the inclusion of phonon dispersion which is responsible for the interaction between the dressed exciton and the phonon subsystem (Davydov 1972). We solve (10) in two iteration steps in the vicinity of $\hbar\omega = E_0$, taking in the first approximation the vertex functions $\Gamma_{n_1 n', \mathbf{k}s}(\omega, \omega_1) = 1$, as done in (Moskalenko 1968). Starting from $\Delta_{(0)}(\omega) = 0$, $\gamma_{(0)}(\omega) = 0^+$ and assuming that $\Delta(\omega) \simeq \text{const}$, we find in the first iteration step

$$\begin{aligned} \Delta_{(1)}(\omega) &\simeq \Delta_{(1)}(E_0/\hbar) = -\Delta' = \frac{\hbar}{N} \sum_{\mathbf{k}} \frac{b_n J_{0n}^o(\mathbf{k})}{E_0 - E_n - \hbar\omega_{\mathbf{k}o}}, \\ \gamma_{(1)}(\omega) &= \frac{\hbar\pi}{N} \sum_{\mathbf{k}} J_{00}^a(\mathbf{k}) \{ [1 + n(\hbar\omega_{\mathbf{k}a})] \delta(\hbar\omega - E_0 - \hbar\omega_{\mathbf{k}a}) + n(\hbar\omega_{\mathbf{k}a}) \delta(\hbar\omega - E_0 + \hbar\omega_{\mathbf{k}a}) \}. \end{aligned} \quad (14)$$

Here b_n denotes the n -th level degeneracy rank ($b_n = 1, 2$). The optical phonon contribution to $\gamma(\omega)$, is significant for $\hbar\omega \simeq E_n \pm \hbar\Omega$ only. We include the LO phonon influence on $\gamma(\omega)$, introducing an effective constant to $J_{00}^a(\mathbf{k})$, which does not influence the time of fast dressing with phonons but it determines the relaxation time at non zero T . This

constant will be estimated from the sum rule $|A(t=0)| = 1$ for $T = 0$. In the second step, the relation $\Delta_{(1)}(\omega) \gg \gamma_{(1)}(\omega)$ enables one to neglect the $\gamma_{(1)}(\omega)$ in the denominators of (10). It leads to

$$\Delta_{(2)}(\omega) \simeq \Delta_{(2)}(E_0/\hbar) = -\Delta'' \simeq \Delta_{(2)}(\tilde{E}_0/\hbar) + \hbar^{-1} \frac{d\Delta_{(2)}}{d\omega} \Big|_{\omega=\tilde{E}_0/\hbar} \Delta' = -\Delta' Z^{-1},$$

$$\gamma_{(2)}(\omega) = \gamma_{(1)}(\omega + \Delta'/\hbar), \quad (15)$$

where $\tilde{E}_n = E_n - \Delta'$, $\Delta_{(2)}(\tilde{E}_0/\hbar) = \frac{\hbar}{N} \sum_{\mathbf{k}} \frac{b_n J_{0n}^o(\mathbf{k})}{E_0 - E_n - \hbar\omega_{\mathbf{k}o}} = -\Delta'$. Continuing the iteration process we would arrive to a self consistent equation for Δ similar to (12), however, after infinite number of the approximations. We find

$$\gamma_{(2)}(x) = Z^{-1} \alpha x^3 e^{-\beta x^2} f(x) \{-n(-x)\theta(-x) + [1 + n(x)]\theta(x)\} \quad (16)$$

where $x = \hbar\omega - E_0 + \Delta'$, $f(x) = \sum_{n=0}^{\infty} \frac{[-\beta x^2 (l_z^2/l_{\perp}^2 - 1)]^n}{(2n+1)n!}$, Z denotes the vertex function renormalisation factor ($Z = |1 - \hbar^{-1} d\Delta/d\omega|^{-1}$). We write the spectral function in the form

$$A(\omega) = Z \frac{\hbar 2\tau^{-1}(\omega)}{[\hbar\omega - E_0 + Z^{-1}\Delta']^2 + \tau^{-2}(\omega)}, \quad (17)$$

where $\tau^{-1}(\omega) = Z\gamma(\omega)$ (Langreth 1964, Mahan 1966). Since at zero temperature, the renormalisation factor may be calculated directly

$$Z \approx 1 + \frac{\hbar}{N} \sum_{\mathbf{k}} \frac{b_n J_{0n}^o(\mathbf{k})}{(E_0 - E_n - \hbar\omega_{\mathbf{k}o})^2}, \quad (18)$$

the sum rule $|A(t=0)| = 1$ enables us to estimate the effective damping constant α . At higher temperatures, when the phonon dispersion effects are strong we evaluate Z using the same condition ($|A(t=0)| = 1$).

3 Results and conclusions

We have evaluated the interaction form factors $J_{nn'}^s(\mathbf{k})$ taking the function of the non-interacting electron and hole system as the first approximation for the exciton wave function $\Phi_n(\mathbf{r}_e, \mathbf{r}_h) = \Psi_{n_e m_e}^{(e)}(\mathbf{r}_e) \Psi_{00}^{(h)}(\mathbf{r}_e)$, where

$$\Psi_{nm}^{(i)}(\mathbf{r}) = \psi_{\perp nm}^{(i)}(r_{\perp}, \varphi) \phi^{(i)}(z) = \frac{N_{nm}^{(i)}}{(2\pi)^{1/2}} \left(\frac{r_{\perp}^2}{l_i^2} \right)^{|m|/2} e^{-\frac{r_{\perp}^2}{2l_i^2}} L_n^{|m|} \left(\frac{r_{\perp}^2}{l_i^2} \right) e^{im\varphi} \frac{1}{\pi^{1/2} l_{iz}} e^{-\frac{z^2}{2l_{iz}^2}},$$

$$N_{nm}^{(i)} = \frac{1}{\pi^{1/2} l_i} \left(\frac{n!}{(n + |m|)!} \right). \quad (19)$$

Since $\omega_0^i \ll \omega_z^i$, we consider only the first level for the quantisation in z-axis direction. A fact that an effective mass of a hole is large in comparison to the electron effective mass, enables one to neglect the hole levels different than the lowest one (of $n_h = 0$, $m_h = 0$). Defining the functions

$$I_{n'n''}^{(i)}(\mathbf{k}) = \int_0^{\infty} \int_0^{2\pi} \psi_{n''}^{(i)*}(r_{\perp}, \varphi) e^{ik_{\perp} r_{\perp} \cos(\varphi)} \psi_{n'}^{(i)}(r_{\perp}, \varphi) dr_{\perp} d\varphi$$

$$\times \int_{-\infty}^{\infty} \phi^{(i)*}(z) e^{ik_z z} \phi^{(i)}(z) dz = I_{\perp n' n''}^{(i)}(k_{\perp}) e^{-2(k_z l_{iz}/2)^2}, \quad (20)$$

where the index n replaces both n_e, m_e quantum numbers, we write the form factors in the form

$$\begin{aligned} J_{nn'}^o(\mathbf{k}) &= \frac{2\pi e^2 \hbar \Omega}{v \tilde{\epsilon} k^2} [I_{nn'}^{(e)}(\mathbf{k}) - I_{nn'}^{(h)}(\mathbf{k})]^2, \\ J_{nn'}^a(\mathbf{k}) &= \frac{\hbar k}{2Mc_a} [\sigma_e I_{nn'}^{(e)}(\mathbf{k}) - \sigma_h I_{nn'}^{(h)}(\mathbf{k})]^2. \end{aligned} \quad (21)$$

We perform the calculations of the energy shift including ten lowest exciton energy levels, for the two confinement sizes $l_e = 6\text{nm} \approx l_h$ and $l_e = 3\text{nm} \approx l_h$. The following material parameters suitable to the InAs/GaAs QD have been used $m_e^* = 0.067m_0$, $m_h^* = 0.38m_0$, $\epsilon_0 = 12.9$, $\epsilon_\infty = 10.9$, $\hbar\Omega = 36.4\text{meV}$, $c_a = 4.8 \cdot 10^3\text{m/s}$. The integral form factors of the exciton-LO phonon interaction $J_{0n}^o = (\hbar/N) \sum_{\mathbf{k}} J_{0n}^o(\mathbf{k})$ multiplied by the level degeneracy rank corresponding to the exciton energies E_n are presented in the table.

The inclusion of many exciton levels influences the dressed exciton energy shift. It is responsible for its substantial increase (from 0.5meV found in one level approximation to 3.6meV when include ten levels for $l_e = 3\text{nm}$, and from 0.35meV to 2.9meV for $l_e = 6\text{nm}$). The other material constants $\sigma_e = 6.7\text{eV}$, $\sigma_h = 2.7\text{eV}$, $\rho = 5.36\text{g/cm}^3$ (Adachi 1985) are useful for estimations.

The spectral density and its inverse Fourier transform calculated including many exciton levels and including realistic asymmetry of the QD ($l_{ze}/l_e = 1/3$) are plotted in Figs 1,2. Generally, the LA channel of dressing gives for the typical QDs the *picosecond scale of dressing*. Inclusion of the LO channel does not modify significantly the overall LO and LA dressing kinetics in comparison to LA channel solely (Takagahara 1999). Thus the LA channel of dressing gives the limit for adiabatic creation of exciton-polaron in InAs/GaAs QD. The slope of the time courses of spectral functions is related to damping of the dressed exciton at non zero temperature. The relaxation time is strongly influenced by the size of QD.

Let us emphasise that our results for time dependence of the response functions (Fig. 1,2) coincide with the experimental results by Borri (2001).

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Figure captions:

Fig. 1. Time course of the response function and spectral intensity for $l_e = 3\text{nm}$, $l_z/l_e = 1/3$

Fig. 2. Time course of the response function and spectral intensity for $l_e = 6\text{nm}$, $l_z/l_e = 1/3$

Table caption:

Exciton energy levels E_n and corresponding integral form factors J_{0n}^o multiplied by n-th level degeneracy rank b_n .

$l_e = 3\text{nm}$		$l_e = 6\text{nm}$	
$E_n[\text{meV}]$	$b_n J_{0n}^o[\text{meV}^2]$	$E_n[\text{meV}]$	$b_n J_{0n}^o[\text{meV}^2]$
88.9	13.1	7.87	12.5
113.3	2· 39.0	15.7	2·18.8
134.0	36.0	23.3	21.5
135.8	2· 16.9	23.8	2·9.9
152.0	2· 18.4	29.6	2·10.4
155.6	2· 5.6	30.8	2·3.5
167.9	18.2	34.8	7.4
169.4	2· 10.7	35.5	2·4.7
183.8	2· 13.0	39.5	2·5.4
199.0	11.5	44.2	4.0

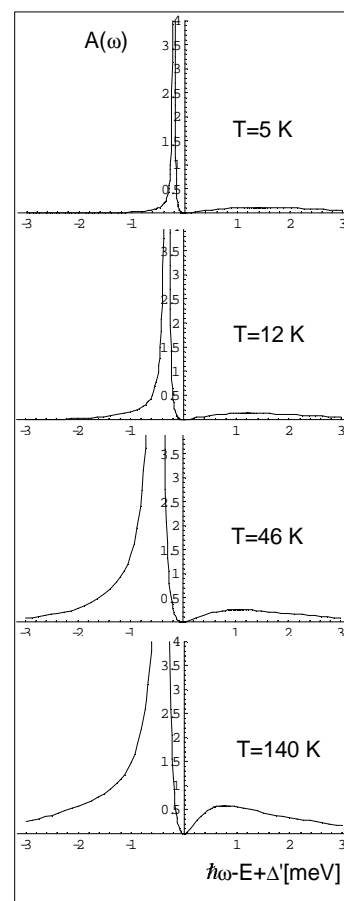
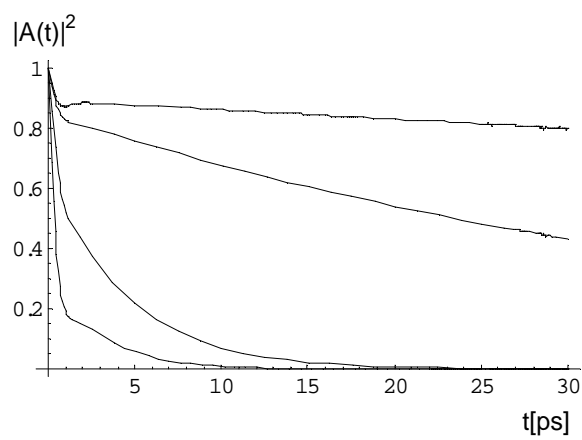


Figure 1

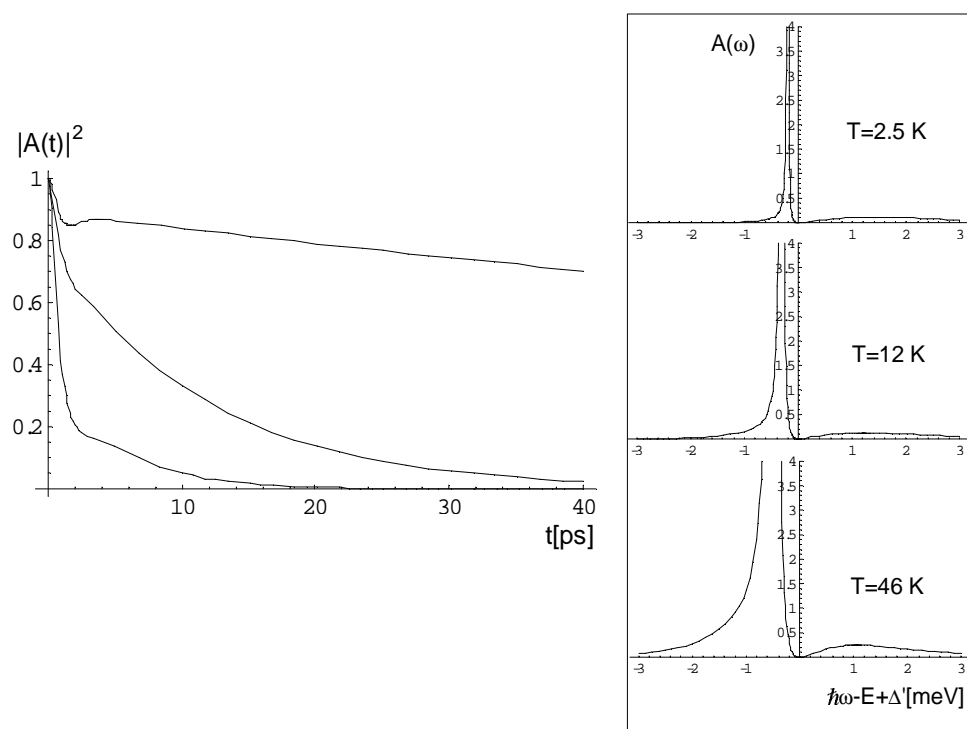


Figure 2